

Site-selective spectroscopy and upconversion processes in $\text{Pb}_5\text{Al}_3\text{F}_{19}$: Nd^{3+} crystal

J. Fernández^{a,b}, R. Balda^{a,b}, M. Sanz^a, A. Mendioroz^a,
^aDpto. Física Aplicada I y ^bCentro Mixto CSIC-
UPV/EHU, Escuela Superior de Ingenieros, Alda.
Urquijo s/n 48013 Bilbao (Spain)
email: wupferoj@bi.ehu.es

J.P. Chaminade and J. Ravez
Institut de Chimie de la Matière Condensée de Bordeaux
(I.C.M.C.B.- C.N.R.S.)
87 Av du Dr Albert Schweitzer 33608 Pessac cedex
(France)

Recently, there has been an interest renewal in solid state lasers, due to great advances in the development of semiconductor laser diodes, which can be used as pumping sources. Upconversion rare-earth solid-state lasers offer potentially simple and compact sources of visible coherent light. Among crystalline lasers, those based on fluoride matrices have been studied intensively in the last two decades [1]. The main peculiarities of the luminescence of rare earth activated fluoride crystals if compared with oxygen-containing compounds are the weak crystal field and weak non-radiative probabilities, resulting in long excited state lifetimes and low concentration quenching.

In this context, $\text{A}_5\text{B}_3\text{F}_{19}$ compositions can provide a large variety of novel insulating crystalline materials [2]. From these compounds we chose the $\text{Pb}_5\text{Al}_3\text{F}_{19}$ composition because it is possible to grow it as a single crystal with appropriate dimensions.

In this work, we have characterized the optical properties of Nd^{3+} ions in fluoroaluminate $\text{Pb}_5\text{Al}_3\text{F}_{19}$ crystals by using steady-state and time-resolved site selective laser spectroscopy. The absorption and emission spectra of Nd^{3+} in this crystal are characterized by inhomogeneously broadened lines similar to those found in glass materials. The features displayed by the emission and excitation spectra, obtained by exciting resonantly the $^4\text{F}_{3/2}$ multiplet with a Ti-sapphire ring laser in the near infrared, 800-920 nm range, indicate the presence of two main distinguishable crystal field centers. Figure 1 shows the low temperature emission spectra obtained by exciting at two different wavelengths. As can be seen, the features of both spectra clearly show the presence of two different centers; however, due to the overlapping between the emissions coming from both main Nd^{3+} sites it is somewhat difficult to know their precise nature and distribution.

The lifetimes of the $^4\text{F}_{3/2}$ level have been obtained at different temperatures and excitation wavelengths along the $^4\text{I}_{9/2} \rightarrow ^4\text{F}_{3/2}$ absorption band. The experimental decays are well described by an exponential function at all temperatures and wavelengths, to a good approximation. The lifetimes are nearly independent of temperature in the 77-295 K range. As we could expect, if different sites for the rare earth were present, the lifetime values should be dependent on excitation and emission wavelengths. In our case, due to the overlapping between the emissions coming from both main Nd^{3+} sites it is somewhat difficult to give accurate values for the lifetime of the excited state of each site. Measurements performed at different

excitation wavelengths show that the lifetime displays a variation of about 15% around 435 μs .

Infrared to visible and ultraviolet upconversion under continuous wave and pulsed laser excitation have been observed. UV and blue, green, yellow and red emissions are observed with excitation at 796 nm within the $^4\text{F}_{5/2}$ level, which is adequate for diode laser pumping. Figure 2 shows the upconverted fluorescence in the 350-700 nm range by exciting in resonance with the $^4\text{F}_{5/2}$ level. Possible mechanisms for two and three photon excitation processes are suggested.

Acknowledgements

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References

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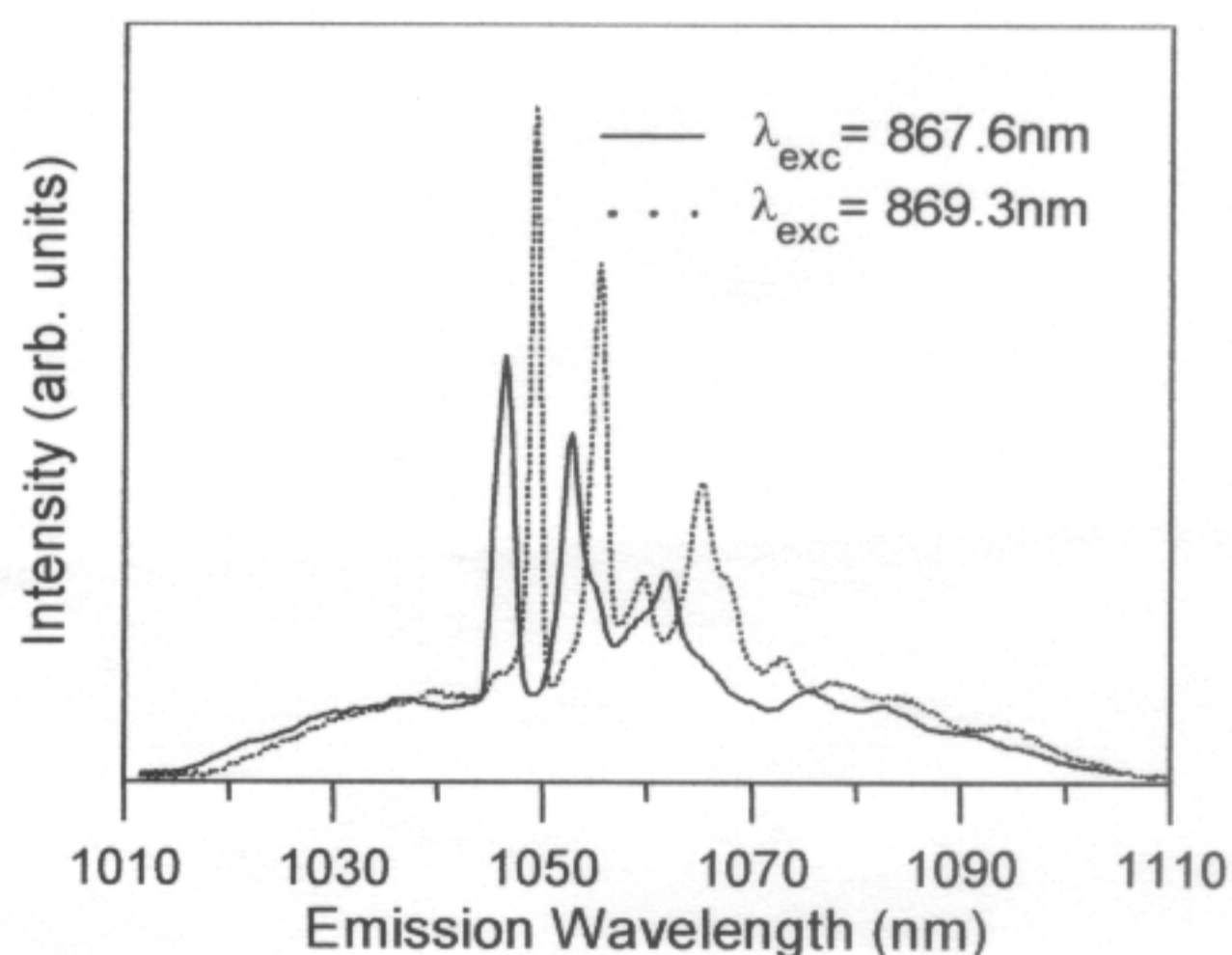


Fig. 1.- Site selective emission spectra for the $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$ transition obtained at 4.2 K.

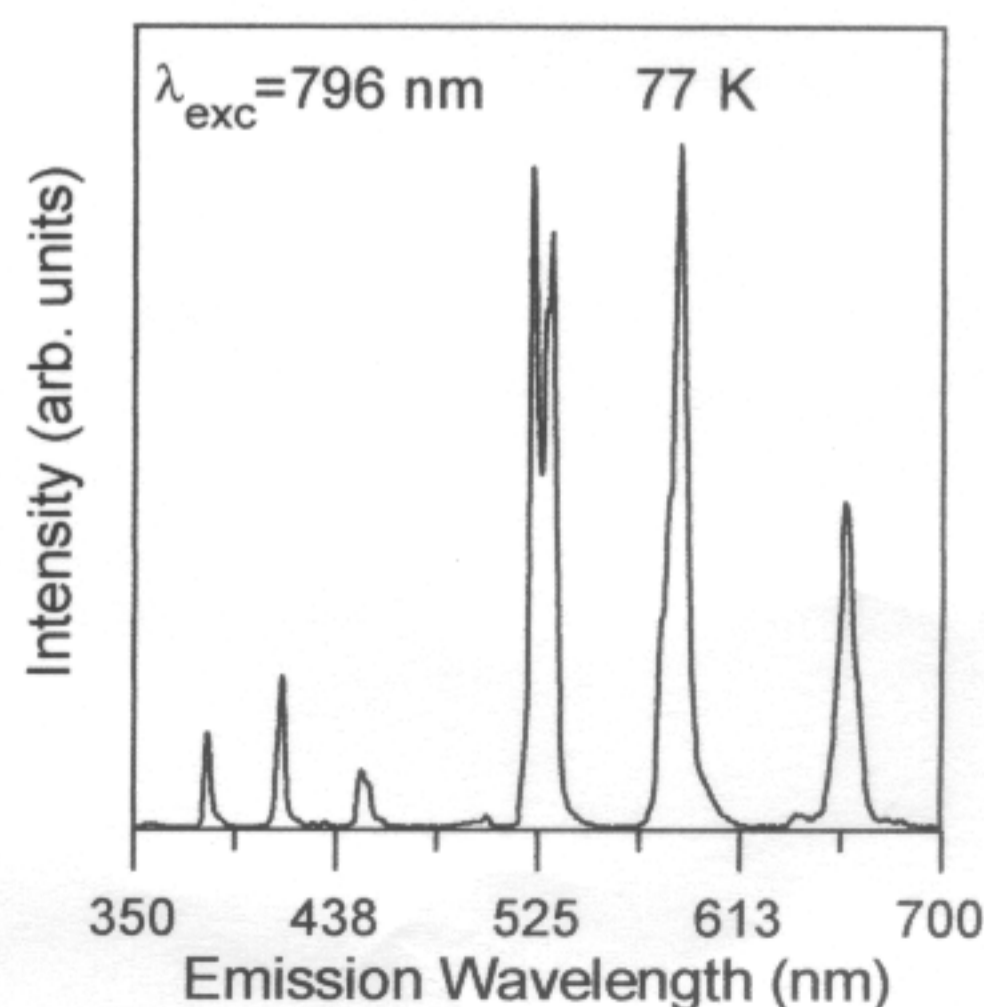


Fig. 2.- Upconversion emission spectrum obtained at liquid nitrogen temperature by exciting in resonance with the $^4\text{F}_{5/2}$ level.